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Bond Strength of Methacrylate-Based Composite to Dentin using Silorane Adhesive

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Bond Strength of Methacrylate-Based Composite to Dentin Using a Silorane Adhesive

ABSTRACT

Objective: The purpose of this study was to compare the shear bond strength of methacrylate-based composite bonded to dentin using a silorane-based adhesive versus other methacrylate-based adhesive bonding agents over time. **Methods**: Eighty human third molars were mounted in PVC pipe, sectioned with a low-speed diamond saw to expose dentin, and surfaces prepared with 600-grit sandpaper. The specimens were randomly divided into four groups of twenty to include a silorane-based adhesive (Filtek LS System Adhesive, 3M ESPE) or one of three methacrylate-based adhesive bonding agents (Clearfil SE Bond, Kuraray; Optibond FL, Kerr; Adper Scotchbond MultiPurpose, 3M ESPE). Adhesive systems were applied according to manufacturer's instructions. Composite (Filtek Z250, 3M ESPE) was inserted into a mold in 2-mm increments to a height of 4 mm and light cured for 20 seconds per increment. Specimens were stored for 24 hours or 6 months in 37°C distilled water, then tested in shear with a universal testing machine (Instron). A mean shear bond strength value (MPa) and standard deviation was determined per group. Data was analyzed with a two-way ANOVA and Tukey's post hoc test (α=0.05). **Results:** No significant difference was found between groups based on bonding agent (p=0.166) or storage time (p=0.219) with no significant interaction (p=0.238). **Conclusions:** Based on this *in vitro* study, it appears that the silorane-based adhesive bonding agent provides comparable shear bond strengths to dentin as proven methacrylate-based adhesive systems and may be used to bond methacrylate-based composite to dentin.

INTRODUCTION

Traditional methacrylate-based composites suffer from volumetric shrinkage during curing, which can lead to polymerization stress and microleakage at the composite-tooth interface. The volumetric shrinkage occurs as a result of the "linear monomers" connecting by shifting closer together in a linear response, resulting in a greater loss of volume. A new silorane-based composite (Filtek LS, 3M ESPE, St. Paul, MN) was recently introduced and differs from traditional methacrylate-based composites in that its ring-opening mechanism decreases polymerization shrinkage significantly. The ring shape of the silorane monomer counteracts the shrinkage by expanding the ring into a linear dimension during the polymerization reaction. The expansion of the ring before polymerization has been shown to decrease the polymerization shrinkage to an average of 1-1.5% (Lien and Vandewalle, 2010). This may produce the desired effect of reduced polymerization stress at the tooth-restorative material interface and less microleakage. 3M ESPE claims that the special silorane chemistry requires a unique adhesive, Filtek LS System Adhesive, when restoring teeth with Filtek LS composite (Filtek LS Technical Profile 2012).

Filtek LS System Adhesive (LSA) is an ultra-mild, two-step, self-etch adhesive system. Self-etch adhesives contain an acidified primer which eliminates the need for a dentin conditioner to remove the smear layer. Self-etch adhesives are classified into one-step (combined acidified primer and adhesive) and two-step systems (acidified primer and separate adhesive) (Van Meerbeek et al., 2010). The manufacturer claims that the LSA adhesive is the only adhesive that can be used with their silorane-based composite restorative material (Filtek LS FAQs, 2012), as it contains a proprietary

hydrophobic bifunctional molecule and an acidic monomer to initiate the ring-opening system.

3M ESPE also states that the LSA may not be used with conventional methacrylate-based composites, as that it will “lead to inadequate bond strengths between the adhesive and the restorative” material (Filtek FAQs, 2012). In seeming contrast, the manufacturer has indicated on their website that Filtek LS composite can be repaired using conventional methacrylate-based adhesives with Filtek LS, or with Filtek LS adhesive and methacrylate-based restorative composites (Filtek LS FAQs, 2012). In 2010, Van Ende and others found that the new silorane system adhesive bonded equally well to dentin using both silorane-based and methacrylate-based (Z100, 3M ESPE) composite restorative materials. Ghulman et al (2011) examined the compatibility of Filtek LS with a methacrylate-based, self-etch adhesive (Adper SE Plus, 3M ESPE.) In their study, they described nanoleakage with water tree formation, and concluded that only silorane adhesives should be used with silorane-based composites. The contradictory nature of these reports suggests that more investigation is needed.

The potential uniqueness of the LSA bonding agent has not received much attention in the literature to date, and its compatibility with other composite resins have not been fully explored. This study examined the shear bond strength of LSA to dentin using Z-250 (3M ESPE), a traditional methacrylate-based hybrid composite, and compared it with several other clinically successful adhesives (Clearfil SE Bond, Kuraray; Optibond FL, Kerr; and Adper Scotchbond MultiPurpose, 3M ESPE) after storage in water for 24 hours and 6 months. See Table 1. For this investigation, long-term water storage was selected as the artificial aging technique to assess long term

bond strength. De Munck noted that “simple ‘water storage’… has a clear bond-degrading effect; it mimics clinically observed restoration degradation very well” (2012).

Two null hypotheses were tested: 1) No significant difference in shear bond strength of methacrylate-based composite to dentin will be found based on type of adhesive bonding agent (silorane or methacrylate based,) or 2) No significant difference in shear bond strength of methacrylate-based composite to dentin using silorane or methacrylate-based bonding agents will be found based on storage time.

METHODS AND MATERIALS

Eighty extracted human third molars were stored in 0.5% chloramine-T at room temperature (24 degrees C) and used within four months of extraction. Third molars that exhibited carious lesions on any surface were excluded from the study. Each third molar was cleaned using scalers and pumice and embedded in Type V dental stone (Die Keen, Heraeus Kulzer, South Bend, IN) and self-cured bis-acrylic resin (Integrity, Dentsply, Milford, DE) to 2mm below the CEJ in a custom cylindrical block. A diamond saw (Isomet, Buhler, Lake Bluff, IL) was used to section the crowns of the teeth horizontally in a mesio-distal direction at the height of contour. The surfaces were examined to ensure complete exposure of the dentin surface and then finished with 10 uniform strokes along 600-grit silicone-carbide sandpaper to create a uniform smear layer.

The teeth were divided into four groups of twenty: 1) Filtek LS System Adhesive (LSA); 2) Clearfil SE Bond (CSE); 3) Adper Scotchbond MultiPurpose (SBMP); and 4) Optibond FL (OFL). Each group had two subsets of 10 teeth for analysis of shear bond

strength at 24 hours and 6 months after bonding. The bonding steps for each of the adhesive system groups listed above were performed according to manufacturer's instructions. Light curing of each adhesive was conducted with a Bluephase 16i light curing unit (Ivoclar Vivadent, Amherst, NY) as recommended by the manufacturers' instructions. Irradiance of the curing light was monitored with a radiometer (LED Radiometer, Kerr) to verify irradiance levels of at least 1200 mW/cm^2.

The specimens were then placed in an Ultradent jig and secured beneath a white, non-stick Delrin mold (Ultradent, South Jordan, UT). The mold standardized the bonding area to a 2.4mm diameter circle. Filtek Z250 composite resin shade A2 was then applied to each specimen in 2-mm increments to a height of approximately 4 mm. Each composite increment was cured for 20 seconds using the light curing unit. The specimens were then stored for their prescribed amount of time (24 hours, 6 months) in distilled water at 37 degrees Celsius.

Upon reaching the respective storage times, each sample was loaded perpendicularly in a universal testing machine (Instron, Norwood, MA) with a crosshead speed of 1.0mm/min. Shear-bond strength values in Megapascals (MPa) were calculated based on the peak load of failure (Newtons) divided by the specimen surface area. The mean and standard deviation was then determined for each group.

Shear-bond strength data was analyzed using a two-way ANOVA and Tukey's post hoc test to evaluate the effects of adhesive bonding agent (4 levels), and time (2 levels) on the shear bond strength of Filtek Z250 composite to dentin at the alpha level of 0.05. The data was further evaluated using two 1-way ANOVAs to compare the bond strength of the adhesive bonding agents at each storage time. Additionally, for each

bonding agent, the bond strength at 24 hours versus 6 months was compared using unpaired t-tests. A Bonferroni correction was applied because multiple comparison tests were done simultaneously (alpha = 0.008).

After completion of testing, each specimen was examined using a stereomicroscope at 10x magnification to determine failure mode. Failure modes were characterized as either 1) adhesive fracture at the composite-adhesive bonding agent interface, 2) cohesive fracture within the composite, 3) mixed failure in the composite or dentin (combined adhesive and cohesive), 4) cohesive fracture within dentin, or 5) adhesive fracture at the adhesive bonding agent-dentin interface.

RESULTS

With the two-way ANOVA, no significant difference in shear bond strength was found between groups based on bonding agent (p=0.166) or storage time (p=0.219) with no significant interaction (p=0.238). With the one-way ANOVAs, no significant difference was found between bonding agents at 24 hours (p=0.248) or at 6 months (p=0.185) of storage in water. The unpaired t-tests found no significant difference per bonding agent between 24 hours and 6 months of storage (p>0.125). See Figure 1.

Although there was no significant difference in bond strength to dentin between the adhesive bonding agents to dentin after 6 months of storage in water, the majority of failures using Clearfil SE Bond were mixed or cohesive in nature suggesting a more stable adhesive interface (Al-Salehi and Burke, 1997). However, the failure mode of LSA was entirely adhesive and similar to the other methacylate-based adhesives,

Optibond FL and Adper Scotchbond MultiPurpose, which were primarily adhesive after 6 months of storage. See Figure 2.

DISCUSSION

Traditional methacrylate-based adhesive bonding agents have changed dramatically in the last several years. Manufacturers have developed new adhesives that are easier and faster to place. Nonetheless, simplification does not always guarantee equal or improved effectiveness (Peumans et al., 2005). Current methacrylate-based adhesives may be divided into two major categories based on the number of clinical steps and their interaction with the tooth surface: etch-and-rinse and self-etch. Etch-and-rinse adhesives may be subcategorized into three- and two-step systems, while self-etch adhesives may be subcategorized into two- and one-step systems (Van Meerbeek et al., 2003). The two-step etch-and-rinse and one-step self-etch are also referred to as simplified adhesives because the primer and adhesive are combined. See Figure 3. The combination of the hydrophilic primer and hydrophobic bonding agent in simplified adhesives has been problematic in the past. The hydrophilic combination acts like a semi-permeable membrane, enabling the transudation of water from the underlying dentin across an osmotic gradient toward the oxygen-inhibited adhesive bonding agent interface, resulting in a reduction in the bond between the adhesive and the overlying composite resin restorative material. In addition, the hydrophilic nature of the hybrid layer in simplified adhesives promotes hydrolytic degradation after long-term exposure to intra-oral fluids (Tay et al, 2004).

A recent meta-analysis evaluating the clinical performance of Class V restorations found that non-simplified adhesive systems that belong to the subcategories of two-step self-etch, such as Clearfil SE Bond, or three-step etch-and-rinse, such as Optibond FL and Adper Scotchbond MultiPurpose, performed better than simplified adhesive systems. More specifically, four independent clinical studies have confirmed the superior performance of Clearfil SE Bond (Heintze et al., 2010). Clearfil SE Bond, the gold-standard of dentinal adhesion, is considered a mild two-step self-etch adhesive similar to the LSA adhesive tested in this study.

LSA primer has been shown to be capable of dissolving calcium ions and binding to apatite surfaces and forming a distinct nano-interaction zone of 100-200 nm, typical of ultra-mild self-etch adhesives such as Clearfil SE Bond (Gregoire et al., 2010; Mine et al., 2010). In the nano-interactive zone, the smear layer is not removed, and the interaction with dentin is superficial, and the residual hydroxyapatite remains available for chemical interaction. This chemical interaction is more stable in an aqueous environment, and occurs between specific monomers and the calcium of hydroxyapatite (Sarr et al., 2010). The additional benefit of a chemical bond may play a role in the shear bond strength, and promote stability of the hybrid layer (Gregoire et al., 2010; Van Meerbeek et al., 2010). The bonds in a mild self-etch adhesive vary greatly, and bond strengths are thought to be related specifically to composition, and to the actual functional monomer in the adhesive formation. The Adhesion-Decalcification Concept model suggests that it is not necessarily the degree of acidity that affects the bond strength, but instead the formation of a stable bond in aqueous solution between the monomer and the calcium of the hydroxyapatite (HAp) crystal (Van Meerbeek et al.,

2010). Van Meerbeek suggests that monomers such as 10-methacryloyloxydecyl dihydrogen phosphate (10-MDP) found in Clearfill SE Bond and polyalkenoic acid in glass ionomers have very limited dissolution of HAp, and the HAp may protect the collagen from degradation by catalytic enzymes. LSA contains a unique monomer, phosphoric-acid methacryloxy hexylester, not previously seen in other adhesive systems.

The manufacturer claims that LSA cannot be used with conventional methacrylate composites because it will lead to “inadequate bond strengths between the adhesive and restorative” (Filtek FAQs, 2012). In this study, we failed to reject the first null hypothesis. The shear bond strength between the silorane adhesive (LSA) and a conventional methacrylate-based composite was comparable to those found with top performing non-simplified methacrylate-based adhesives (OFL, SBMP, CSE). This agrees with a study by Van Ende and others (2010) who found similar bonding to dentin with a methacrylate-based composite using either a silorane- or methacrylate-based adhesive.

We also failed to reject the second null hypothesis. No significant difference in shear bond strength was found between the groups based on storage time. Although there was no significant differences in bond strength between the adhesive systems, Clearfil SE Bond had more mixed failures after 6 months of storage in water compared to LSA and the other methacrylate-based adhesives, which had primarily adhesive failures. MDP, the functional monomer in Clearfil SE Bond, has been shown in laboratory studies to provide chemical adhesion in addition to the traditional micro-

mechanical adhesion to dentin as found with other methacrylate-based bonding agents (Van Meerbeek et al., 2010) and may partially explain the differences in failure mode.

A limitation to this study is that only one composite resin material was studied. Different results may have been found using other methacrylate-based composites due to variations in formulation. Relatively high standard deviations were found with some of the groups, especially after 6 months of storage time. The higher standard deviations may be associated with the variability in the level of hydrolysis through the hybrid layer over time. Also, the specimens were only stored for six months in water. Longer storage times may produce greater degradations in bond strength of the adhesive bonding agents.

Although more research is necessary, the results of this laboratory study would suggest that practitioners using Filtek LS in their clinic may consider using LSA for bonding both Filtek LS and methacrylate-based restorative composites to dentin.

CONCLUSION

Based on this *in vitro* study, the type of adhesive bonding agent (silorane- or methacrylate-based) did not have a statistically significant effect on the shear bond strength of a methacrylate-based composite resin restorative material to dentin after 24 hours or 6 months of storage in water. The silorane-based adhesive bonding agent, Filtek LS System Adhesive (LSA), appeared to demonstrate comparable shear bond strengths as other methacrylate-based systems tested and appears to be adequate to bond the methacrylate-based composite, Filtek Z250, to dentin.

DISCLOSURE

The views expressed in this study are those of the authors and do not reflect the official policy of the United States Air Force, the Department of Defense, or the United States Government. The authors do not have any financial interest in the companies whose materials are discussed in this article.

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Table 1- Study Materials

Material	Type	Manufacturer	Resin	Filler
Filtek Z250	Hybrid methacrylate-based composite resin	3M/ESPE St. Paul, MN	Bis-GMA, Bis-EMA, UDMA, TEGDMA	Zirconia, silica
LS System Adhesive	2-step self-etch methacrylate-based bonding agent	3M/ESPE St. Paul, MN	HEMA; Bis-GMA	Silane-treated Silica
Clearfil SE Bond	2-step self-etch methacrylate-based bonding agent	Kuraray, New York, NY	HEMA; Bis-GMA	Silanated colloidal silica
Adper Scotchbond Multi-Purpose	2-step etch-and-rinse methacrylate-based bonding agent	3M/ESPE St. Paul, MN	HEMA; Bis-GMA	N/A
Optibond FL	2 step etch-and-rinse methacrylate-based bonding agent	Kerr Orange, CA	HEMA; Bis-GMA	Silica

Figure 1- Graph of Shear Bond Strength

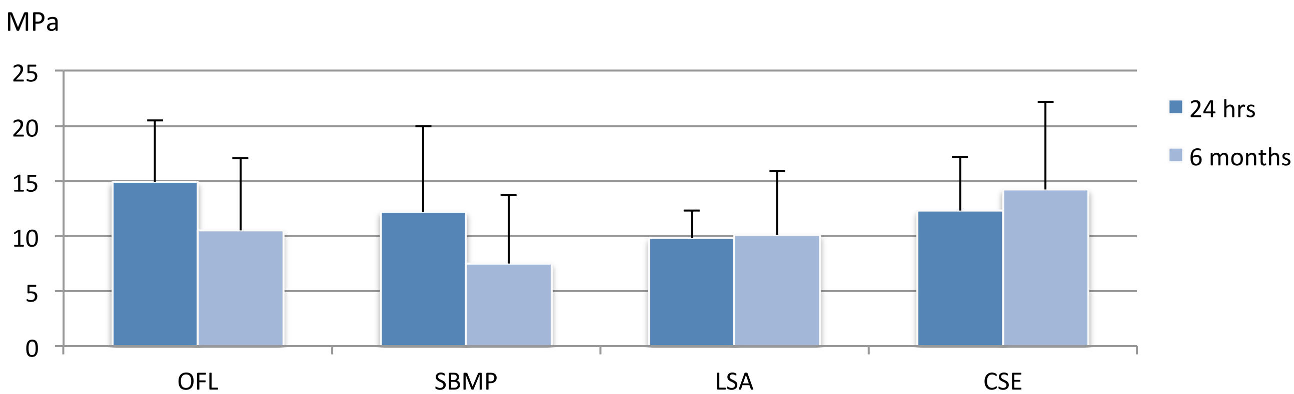

Figure 2- Graph of Failure Modes

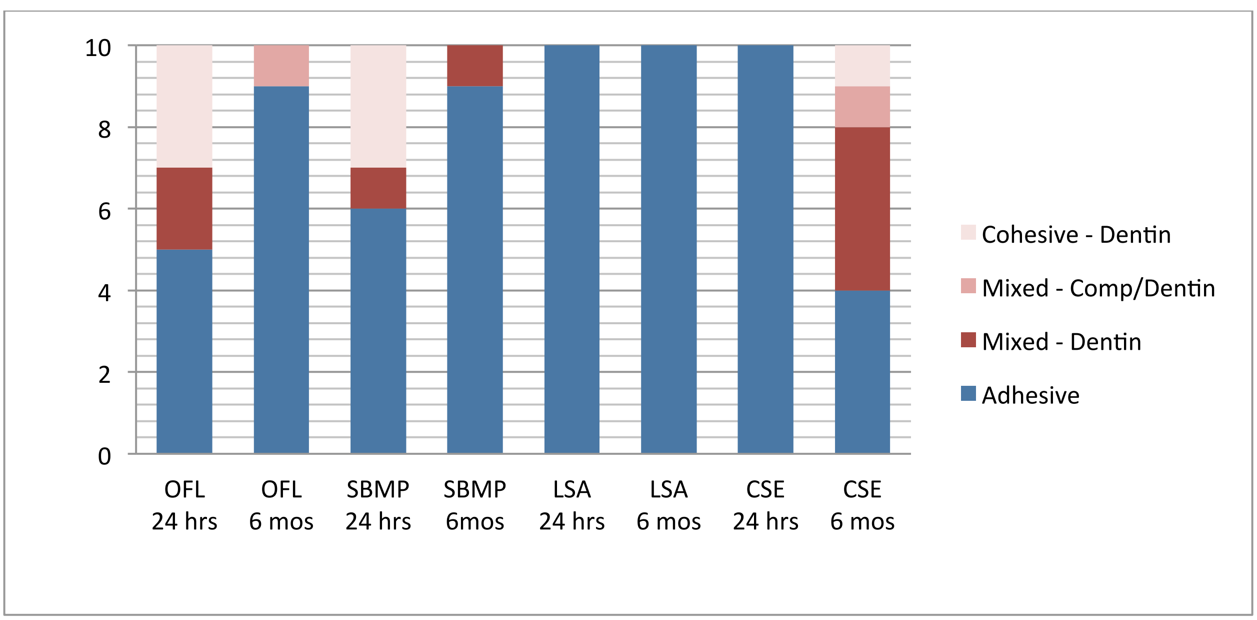

Figure 3 – Adhesive Classifications